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Report C86-14

# ELECTROCHEMICAL HYDROGEN CONCENTRATOR FOR

## PHOSPHORIC ACID FUEL CELLS

Prepared by:

José Giner, Ph.D.
Nancy D. Kackley

GINER, INC.
14 Spring Street
Waltham, MA 02254-9147

November, 1987

Final Report

September 30, 1986 - September 30, 1987

Contract No. DAAK70-86-C-0114

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Ft. Belvoir, VA 22060-5606

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The production of high purity hydrogen for use in a phosphoric acid fuel cell from heavily contaminated reformed diesel fuel gas using an electrochemical device has been demonstrated. The electrochemical hydrogen separator (EHS) is based on current PAFC technology. Hydrogen is removed from the contaminated gas stream by oxidation to hydrogen ions at a gas diffusion anode and recombination at a gas diffusion cathode. An anode catalyst with excellent tolerance to high levels of CO and $H_2S$ contaminants has been developed. The use of this anode catalyst would allow the EHS device to produce pure hydrogen with minimal voltage impact on the overall PAFC power output. Reduction in size and weight of the PAFC power plant is also indicated through reduced load on fuel processing components.

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## EXECUTIVE SUMMARY

The phosphoric acid fuel cell (PAFC) requires hydrogen feed gas at the anode which is low in contaminants such as carbon monoxide and hydrogen sulfide. These contaminants will poison the typical PAFC anode for hydrogen oxidation. Therefore, in order to utilize a logistic fuel such as diesel gas in the PAFC, extensive gas cleanup is necessary. However, the fuel processing components add excessive weight and volume to a PAFC power generator system. Simplification of the fuel processing system through the development of an electrochemical hydrogen separator (EHS) was addressed in this program. The EHS electrochemically consumes hydrogen gas from a heavily contaminated fuel gas at the anode, providing essentially pure hydrogen fuel at the cathode for subsequent use in the PAFC.

The experimental goals of this program included the identification of the best anode catalyst for the EHS through parametric testing of six candidate catalysts at varied cell temperatures, current densities and $H_2$ gas utilizations. The performance goal was to minimize the operating voltage of the EHS by developing a catalyst with high activity for the hydrogen oxidation reaction as well as tolerance for contaminant gases such as CO and $H_2S$. In addition, the $H_2S$, CO and $CO_2$ content of the product gas was measured. The catalysts and electrodes were analyzed by elemental analysis, transmission electron microscopy, and X-ray diffraction. Catalyst surface area was measured by electrochemical hydrogen adsorption using cyclic voltammetry. Testing for the water-gas shift reaction or direct CO oxidation was also performed. Generation of additional hydrogen from the high CO content of reformed diesel fuel gas by the shift reaction or direct oxidation of CO to $CO_2$ by $H_2O$ would increase the energy content of the EHS product gas.

An efficient anode catalyst for the EHS was identified. This catalyst was able to operate at low cell voltages at 210°C, 230°C and 250°C on simulated reformed diesel fuel gas (SRDFG) based on

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the output of the Ralph M. Parsons' Co. reformer. This SRDFG contained 49% $H_2$, 16% CO, 11% $CO_2$, 24% $N_2$ and 800 ppm $H_2S$. The catalyst showed tolerance for these high contaminant concentrations. Performance of the EHS cell on Parsons' SRDFG was possible at only 14 mV above the operating voltage on pure hydrogen at 230°C, 70% gas utilization, 200 mA/cm$^2$. The product gas contained only 6 ppm $H_2S$, 0.3% CO and 0.6% $CO_2$. This catalyst showed good potential for allowing the EHS device to efficiently produce pure hydrogen from heavily contaminated diesel fuel gas for use in the PAFC.

The benefits of using this device are reduced weight and volume of the fuel processing unit with minimal loss in overall output voltage of the PAFC power generating plant. Complete elimination of the shift convertor components of the fuel processing unit is probably not feasible since a large portion of the energy value of the SRDFG is in the CO content of the gas. However, the increased tolerance to CO of the EHS anode would permit reductions in the size and weight of the shift convertor catalyst bed. An increase in CO content of the post-shift reformer gas from 2% to 5% could be tolerated by use of the EHS with the advanced anode catalyst identified in this program. This would allow an estimated 10 to 20% reduction in size and weight of the shift reformer. In addition, since tolerance to high levels of $H_2S$ has been demonstrated with this catalyst, the ZnO guard bed may also be reduced in size or eliminated, resulting in additional reduction of the fuel processing system size and weight.

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## 1.0 INTRODUCTION

The overall objective of this program was to develop an electrochemical hydrogen concentrator for use in the fuel processing system of a diesel fueled phosphoric acid fuel cell. A schematic diagram of a conventional diesel fueled PAFC is shown in Figure 1. The five separate units perform three basic functions; 1) conversion of the hydrocarbon fuel to $H_2$, $CO_2$ and CO; 2) removal of sulfur-based contaminants; and 3) removal of CO through shift conversion with water to $H_2$ and $CO_2$. These units are necessary to convert the diesel fuel to a purified hydrogen-rich gas stream for direct use in a phosphoric acid fuel cell. The PAFC anode is easily poisoned by contaminant gases such as CO and $H_2S$ (1). These species, when adsorbed on the platinum catalyst surface, reduce the number of available sites for the hydrogen oxidation reaction which reduces the overall power output of the fuel cell. Dilutant gases, such as $N_2$ and $CO_2$, also decrease the fuel cell efficiency by reducing the amount of hydrogen available for utilization (2).
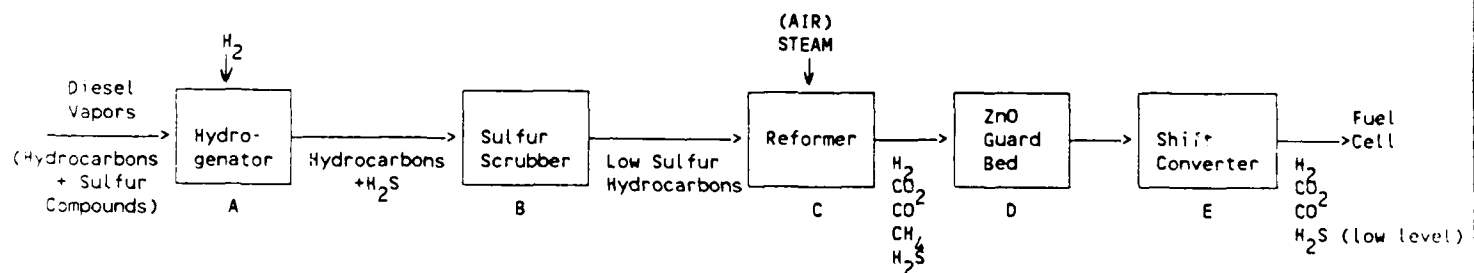


A: Hydrogenator
B: Sulfur Scrubber
C: Reformer
D: Sulfur (ZnO) Guard bed
E: Shift converter

Figure 1  Block Diagram of a Conventional PAFC Fuel Processing Train

While the PAFC can be a lightweight, compact power generating unit, the fuel processing components necessary to convert a logistic fuel like diesel into a useable PAFC fuel gas add excessive weight and volume. Simplification of the fuel processing system to three basic units; reformer, sulfur scrubber and hydrogen

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concentrator was one approach identified by the U.S. Army Belvoir RD&E Center. This program has addressed the development of a hydrogen concentrator unit, specifically, an Electrochemical Hydrogen Separator (EHS). The EHS produces essentially pure $H_2$ from a contaminated feed gas by electrochemical removal of the hydrogen from the gas stream. The product gas can be fed directly into a PAFC.

The EHS device is based on phosphoric acid fuel cell technology. A major difference, however, is that hydrogen-based gases are present at both the anode and the cathode. Hydrogen is removed from a contaminated gas stream by oxidation to hydrogen ions at a gas diffusion anode. The hydrogen ions are then electrochemically transported across the acid electrolyte matrix and are recombined into hydrogen at a gas diffusion cathode. Since current must be supplied to perform the separation, the unit operates at a voltage polarized slightly away from zero volts due to internal resistance and some activation polarization.

The absence of an air or oxygen cathode in the EHS cell provides three primary benefits important to this project: 1) due to the absence of oxygen, operation at temperatures higher than normally used in PAFC cells can be investigated without causing corrosion of the cathode material, 2) the cathode functions essentially as a reproducible hydrogen electrode, with minimal polarization, which allows even small decreases in anode polarization due to improved catalysts to be visible, and 3) experimental safety is increased as volatile hydrogen-oxygen gas mixtures are not present.

The polarization of the EHS cell, and therefore the power requirement, can be minimized by choosing the optimum operating conditions and using the most efficient anode catalyst. Six anode catalyst formulations supported on high surface area carbon were tested under an extensive matrix of conditions. Performance of the various catalysts was tested using pure hydrogen gas and a simulated reformed diesel fuel gas. Performance on pure hydrogen

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gas feed to the anode was considered a baseline, as the contaminant and dilutant effects of the SRDFG gas mixture were absent.

In the majority of the work performed, the SRDFG gas composition was based on the output of the Ralph M. Parsons Co. two-stage diesel reformer (3). (In some limited preliminary testing, an SRDFG mixture based on the International Fuel Cell diesel reformer was used.) The Parsons' SRDFG mixture contained 49% $H_2$, 16% CO, 11% $CO_2$, 24% $N_2$ and 800 ppm $H_2S$. Carbon monoxide and hydrogen sulfide are known to be poisons for the hydrogen oxidation reaction on platinum catalysts (1,2). The advanced anode catalysts tested in this program were selected with the goal of developing an efficient, contaminant-tolerant anode catalyst for use in the EHS cell.

Contaminant-induced poisoning of the anode catalyst is indicated by increased cell voltage required to drive the hydrogen separation process. This may lead to decreased current and concomitant decreased hydrogen output at the cathode side of the cell. The performance of various anode catalysts was evaluated by measuring the cell voltage during operation on pure $H_2$ and on the SRDFG mixture under a test matrix of varying cell temperature, gas utilization and current density. The result of this investigation was the development of an EHS cell configuration capable of producing essentially pure hydrogen from a heavily contaminated reformer gas with minimal voltage input requirement. A novel, Pt-alloy anode catalyst has been identified for the hydrogen oxidation reaction which can operate on a feed gas containing high levels of carbon monoxide and hydrogen sulfide with minimal polarization. The experimental details and results are presented below.

## 2.0  EXPERIMENTAL PROCEDURES

### 2.1  EHS Cell Parametric Testing

The EHS cell consisted of two graphite blocks encasing gas diffusion anode and cathode electrodes separated by a SiC matrix. A schematic of the cell is shown in Figure 2. The active
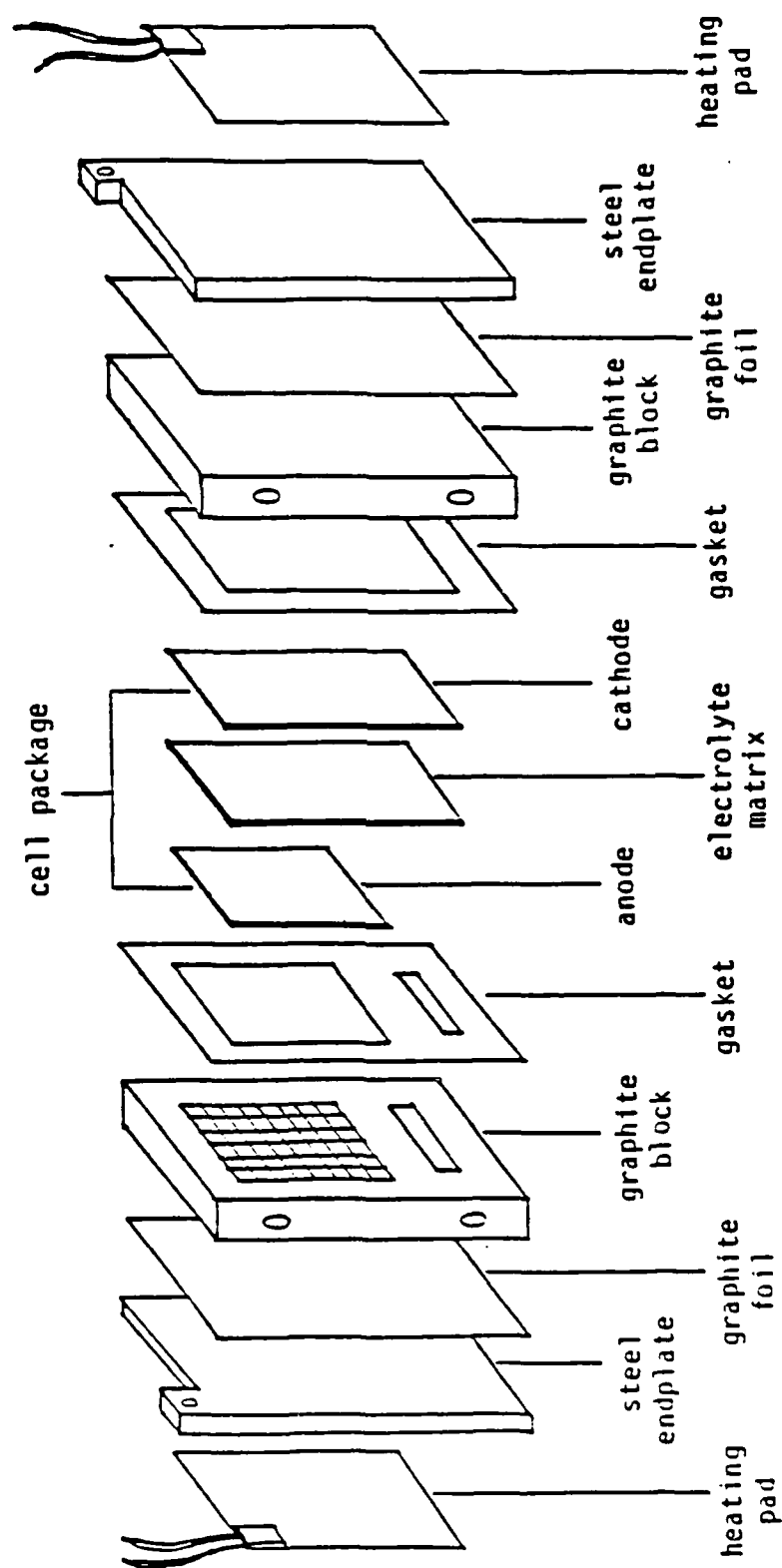
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Figure 2. Schematic of EHS Cell

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cell area was 2" x 2"; approximately 25 cm$^2$. The cathode electrocatalyst was 10% platinum supported on Vulcan XC-72 carbon at a platinum loading of 0.5 mg/cm$^2$. The cathode electrode was formed with 40% PTFE, sintered at 345°C. The anode structure consisted of a Pt or Pt-alloy electrocatalyst on Vulcan XC-72 with 50% PTFE, sintered at 335°C, in all but one case (where 40% PTFE was used). The Pt loading was also 0.5 mg/cm$^2$ on the anode. The details of the anode electrode compositions are presented in the Appendix. The electrolyte, encased in the SiC matrix layer, was 100% $H_3PO_4$.

The testing parameters for evaluation of different anode electrocatalysts included variations in cell temperature, hydrogen utilization, and current density. The parameters used are shown in Table 1. The hydrogen utilization was controlled by adjusting the inlet gas flow rate to the anode at each current density. At each cell temperature and gas utilization, the cell voltage was measured at current densities from 200 to 1000 mA/cm$^2$. The operating voltage of the EHS was measured under the above parameters first using pure $H_2$ gas feed to the anode. Then, the experimental matrix was repeated using a simulated reformed diesel fuel gas. The majority of the testing was done using an SRDFG mixture based on the Ralph M. Parsons Co. reformer (3). Some preliminary tests using only one anode catalyst were performed using a gas mixture based on the International Fuel Cell reformer (4). The compositions of these two gas mixtures are shown in Table 2. The Parsons' SRDFG has much higher CO and $H_2S$ concentrations than the IFC mixture, and constitutes a "worst-case" fuel gas. The CO, $CO_2$ and $H_2S$ content of the cathode exit gas was measured. A small amount of these contaminant gases may cross the cell by diffusion through the electrolyte.

**TABLE 1**

**Parameters varied in EHS testing**

| Parameter | Values |
|---|---|
| Cell Temperature | 190, 210, 230 & 250°C |
| Hydrogen Utilization | 30%, 70% & 90% |
| Current Density | 200, 400, 600, 800, 1000 mA/cm$^2$ |

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## TABLE 2

**Simulated reformed diesel fuel gas mixtures based on Parsons' and IFC reformer outputs**

| Company | Volume % (dry basis) | | | | |
| --- | --- | --- | --- | --- | --- |
| | $H_2$ | $CO_2$ | CO | $N_2$ | $H_2S$ (ppm) |
| Parsons | 49 | 11 | 16 | 24 | 800 |
| IFC | 73 | 21.4 | 5.7 | -- | 180 |

The feed gas was saturated by bubbling through water. This was necessary to maintain the water content of the electrolyte at the elevated cell temperatures. The saturator temperature was normally held at 73°C. However, in later testing, the saturator temperature was raised to 85°C for testing at cell temperatures of 230°C and 250°C. This was done in an attempt to prolong cell life and reduce the cell resistance by increasing the water vapor pressure over the phosphoric acid. The conductivity of phosphoric acid sharply decreases with increasing concentrations above 95% (5). The resistance of the cell package was measured using a current interrupt technique.

The high CO content of the reformed diesel fuel gas can be converted into additional hydrogen through the water-gas shift reaction:

$$CO + H_2O = CO_2 + H_2$$

or direct CO oxidation

$$CO + H_2O = CO_2 + 2H^+ + 2e^-$$
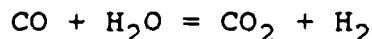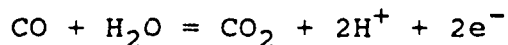
In order to test for the occurrence of either of these reactions at the EHS anode, a gas composition of 24% CO, 76% $N_2$ saturated with $H_2O$ at 73°C was fed to the EHS cell. The amount of hydrogen exiting from the cathode and CO and $CO_2$ content of the anode exit were measured at current densities from 0 to approximately 240 mA/cm². As the current density increased, the

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increasing $CO_2$ content of the anode exit gas indicated that either the shift reaction or direct CO oxidation was occurring. The cell voltage necessary to drive this reaction was also measured. Since this testing was likely to poison catalysts that were not CO-tolerant, it was only performed after all other testing on the cell was completed.

## 2.2   Preparation and Characterization of Catalysts and Electrodes

The Pt-alloy catalysts were prepared by depositing the alloy component onto a platinized high surface area carbon support material. Starting materials were metal salts. Through the addition of hydroxide based dispersing agents, the deposition of small particles of metal hydroxides on the platinized carbon support material was accomplished. The Pt-alloy catalysts were then heat-treated at 900°C in $N_2$ or $H_2$ in order to reduce the hydroxides to pure metals.

Elemental analyses were performed on catalyst preparations to confirm that compositions of Pt-alloy catalysts were as desired. The particle size of the electrocatalysts was determined by transmission electron microscopy (TEM) of the supported catalyst powders.

Pre-test and post-test anodes and cathodes were also examined using transmission electron microscopy. Particles of the supported catalyst were scraped from the electrodes onto TEM grids. The morphology and distribution of catalyst particles before and after testing was compared. Surface areas of the pre-test and post-test anodes and cathodes were measured using an electrochemical hydrogen adsorption technique.

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## 3.0 __RESULTS__

The results presented below are grouped by the type of anode catalyst used in the EHS cell, since other cell components and variables were equivalent for all cells, as discussed in Section 2.1. Evaluation of the EHS cell performance using different anode catalysts was made by comparing the overall cell voltage during operation on the heavily contaminated SRDFG (see Table 2 for composition) and on pure $H_2$. Operation on pure $H_2$ was regarded as a baseline for hydrogen oxidation efficiency of the catalyst, since the contaminant and dilutant gases are not present. Operation on the SRDFG measured the contaminant tolerance of the six anode catalyst formulations. As discussed in Section 1.0, minimization of the EHS cell voltage required to produce pure $H_2$ from SRDFG was a measure of the efficiency of the anode catalyst. In addition, the cathode exit gas was analyzed for trace contaminant content to measure diffusion of these species across the cell package. The results of testing for the water-gas shift reaction or CO oxidation are also presented for each catalyst formulation. Results of pre- and post-test characterization of the catalysts and electrodes are discussed in order to explain differences in performance of the six catalyst formulations.

### 3.1  __10% Pt/Vulcan XC-72__

The first electrocatalyst tested in the EHS cell was 10% Pt/Vulcan XC-72, a commercially available product (Johnson & Matthey, Inc.) often used in phosphoric acid fuel cell anodes. The platinum component had a particle size of 15-25 Å. In initial tests, this anode catalyst was used in an EHS cell with a 4 mil thick, 1500 grit SiC electrolyte matrix. The standard cathode electrode, also 10% Pt/Vulcan XC-72, was used. Performance on pure $H_2$ and on SRDFG based on the International Fuel Cell reformer output was tested in this cell. Another EHS cell with the same 10% Pt/Vulcan XC-72 anode and cathode catalysts and an 8 mil, 1500 grit SiC electrolyte matrix was tested using SRDFG based on the Parsons' reformer output. The voltages of these two cells at 70% $H_2$

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utilization are shown in Table 3. The cell with the 4 mil matrix displayed much lower voltages on pure $H_2$ than the cell with the 8 mil matrix. The difference in the cell resistance correction factor accounted for some, but not all, of this difference in performance. The performance on SRDFG demonstrated the effect of the higher contaminant concentrations present in Parsons' mixture. The increase in cell polarization (difference between voltage on SRDFG and on pure $H_2$) during operation on Parsons' SRDFG indicated the increased poisoning effects due to higher CO and $H_2S$ content.

## TABLE 3

EHS cell voltages using 10% Pt/Vulcan anode catalyst.   Uncorrected cell voltages on $H_2$ and on IFC and Parsons' SRDFG at 70% $H_2$ utilization rate.

| Cell Temp., °C | Current Density mA/cm² | Cell Voltage, mV | | | | | |
|---|---|---|---|---|---|---|---|
| | | 4 mil Matrix | | | 8 mil Matrix | | |
| | | $H_2$ | IFC | $\Delta V$ | $H_2$ | Parsons' | $\Delta V$ |
| 210 | 200 | 27 | 35 | 8 | 76 | Not tested | |
| | 400 | 48 | 74 | 26 | 151 | Not tested | |
| | 600 | 72 | 112 | 40 | 223 | Not tested | |
| | 800 | 95 | 156 | 61 | 298 | Not tested | |
| | 1000 | 119 | ---* | -- | 376 | Not tested | |
| Cell Resistance Correction Factor (ohm-cm²) | | 0.09 | 0.11 | | 0.30 | | |
| 230 | 200 | 27 | 32 | 5 | 88 | 112 | 24 |
| | 400 | 54 | 64 | 10 | 169 | 210 | 41 |
| | 600 | 81 | 99 | 18 | 251 | 303 | 52 |
| | 800 | 108 | 130 | 22 | 342 | --- | -- |
| | 1000 | 134 | 164 | 30 | 429 | --- | -- |
| Cell Resistance Correction Factor (ohm-cm²) | | 0.09 | 0.11 | | 0.32 | 0.30 | |
| 250 | 200 | 33 | 33 | 0 | 98 | 118 | 20 |
| | 400 | 59 | 66 | 7 | 194 | 228 | 34 |
| | 600 | 88 | 100 | 12 | 291 | 327 | 35 |
| | 800 | 132 | 132 | 0 | 392 | 459 | 67 |
| | 1000 | 151 | 163 | 12 | 497 | 600 | 103 |
| Cell Resistance Correction Factor (ohm-cm²) | | 0.09 | 0.11 | | 0.38 | 0.38 | |

*Voltage unstable and rising

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Testing for the shift reaction or direct CO oxidation using a $CO/H_2O/N_2$ gas mixture was not carried out in either of these cells. There was no evidence for conversion of CO to $CO_2$ under normal testing conditions on the SRDFG mixture.

After testing, the surface area of the platinum catalyst from the cell with a 4 mil matrix was measured by hydrogen adsorption using cyclic voltammetry. The pre-test surface area measured 110 $m^2/g$. The anode post-test surface area was 48 $m^2/g$; the cathode measured 51 $m^2/g$. The surface area of the anode from the cell with the 8 mil matrix could not be measured as the electrode was dried out by failure of the saturator. The cathode surface area dropped from 110 $m^2/g$ to 68 $m^2/g$. This reduction in surface area while the cells were on test was due to sintering and agglomeration of the supported Pt crystallites. Since this is a diffusional process, it is enhanced by high temperature operation. However, the surface area loss cannot be attributed to any one aspect of EHS operation, as testing was performed under a wide variety of conditions.

The product gas composition (cathode exit) showed lower contaminant gas contents in the cell with a thicker electrolyte matrix even though a more heavily contaminated SRDFG was used. The EHS cell with an 8 mil SiC matrix averaged 31 ppm $H_2S$, 1% CO and 1% $CO_2$, dry basis in the cathode exit gas. The EHS cell with a 4 mil SiC matrix averaged 35 ppm $H_2S$, 1% CO and 3-4% $CO_2$, dry basis. All other catalyst formulations were tested only in EHS cells with an 8 mil electrolyte matrix due to failures by electrical shorting of EHS cells with a 4 mil matrix.

## 3.2   900°C Heat-Treated 10% Pt/Vulcan XC-72

High surface area platinized carbon materials used in phosphoric acid fuel cells are often heat-treated to improve their performance and long-term stability. Heat treatment of platinized carbon materials also results in an increase in particle size of the catalyst component. The performance of a 900°C heat-treated

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10% Pt/Vulcan XC-72 anode catalyst was investigated in the EHS cell. This catalyst had a particle size of 40-60 Å, approximately 2.5 times that of the non-heat-treated 10% Pt/Vulcan XC-72 catalyst. Testing of this catalyst was performed in an EHS cell with an 8 mil SiC electrolyte matrix and a standard 10% Pt/Vulcan XC-72 cathode. The cell voltages on pure $H_2$ and on Parsons' SRDFG at 230°C and 250°C are shown in Table 4.

## TABLE 4

**EHS cell voltages using 900°C heat treated 10% Pt/Vulcan XC-72 anode catalyst. Uncorrected cell voltages on $H_2$ and on Parsons' SRDFG at 70% $H_2$ utilization rate.**

| Cell Temp., °C | Current Density mA/cm² | Cell Voltage, mV | | |
|---|---|---|---|---|
| | | $H_2$ | SRDFG | $\Delta V$ |
| 230 | 200 | 48 | 67 | 19 |
| | 400 | 96 | 126* | 30 |
| | 600 | 145 | --- | -- |
| | 800 | 194 | --- | -- |
| | 1000 | 242 | --- | -- |
| Cell Resistance Correction Factor (ohm-cm²) | | 0.17 | 0.14 | |
| 250 | 200 | 51 | 75 | 24 |
| | 400 | 100 | 127 | 27 |
| | 600 | 158 | 175 | 17 |
| | 800 | 200 | --- | -- |
| | 1000 | 253 | --- | -- |
| Cell Resistance Correction Factor (ohm-cm²) | | 0.16 | 0.14 | |

*Voltage unstable and rising

The cell voltages for the heat-treated catalyst were consistently lower than for the non-heat-treated catalyst. This indicates that the heat-treated 10% Pt/Vulcan XC-72 was a more efficient catalyst for the hydrogen oxidation reaction, with either pure hydrogen or a heavily contaminated diesel fuel gas. Performance on SRDFG for the heat-treated catalyst at 230°C and 250°C (and the non-heat-treated catalyst at 230°C) was limited to low current densities; at higher current densities, the cell

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voltage was unstable and rising rapidly. The product gas at the cathode of this cell contained 0.4% CO, 0.3% $CO_2$ and 50 ppm $H_2S$ at 600 mA/cm$^2$, 250°C.

The surface area of the 900°C heat-treated catalyst, measured by hydrogen adsorption with cyclic voltammetry, was 60 m$^2$/g before EHS testing. The effect of heat-treatment in increasing the catalyst particle size is evident from comparing this to the pre-test surface area of non-heat-treated 10% Pt/Vulcan XC-72 catalyst; 110 m$^2$/g. However, the post-test surface area of the heat-treated catalyst, 50 m$^2$/g, is about equal to that of the post-test non-heat-treated catalyst. The cathode catalyst displayed a larger decrease in surface area; 110 m$^2$/g, pre-test and 36 m$^2$/g, post-test. The reason for this large decrease in surface area on the cathode is unclear. X-ray analysis of the pre-test and post-test anode electrodes was performed. The peak positions remained essentially the same, but the peak heights were greatly reduced after EHS cell testing. This indicates that some loss of catalyst may have occurred.

Experiments using a $CO/H_2O/N_2$ feed gas indicated that the shift reaction or direct CO oxidation was occurring in this cell at voltages from 550 mV to 650 mV. At 250°C, the amount of $CO_2$ in the anode exit increased from 4% at 80 mA/cm$^2$ to 9% at 200 mA/cm$^2$, while the CO content decreased from 8.5% to 3%. However, the cell voltage increased from 552 mV at 80 mA/cm$^2$ to 650 mV at 200 mA/cm$^2$. Similar voltage levels and $CO_2/CO$ contents were measured at 230°C. The amount of hydrogen produced by this reaction also increased with increasing current density, from 9.75 cc/min at 80 mA/cm$^2$ to 29.7 cc/min at 200 mA/cm$^2$.

## 3.3  Catalyst G86-14-3

In the Phase I portion of this program, this material was identified as an efficient anode catalyst for EHS cell operation on gas containing up to 10% CO and 2000 ppm $H_2S$ (6). Further testing of this catalyst in this Phase II program using Parsons' SRDFG confirmed the prior results. EHS cell voltages and cell

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polarizations (difference between cell voltage on $H_2$ and on SRDFG) using the G86-14-3 anode electrocatalyst are shown in Table 5. When compared to the performance of the heat-treated 10% Pt/Vulcan electrocatalyst (Table 4), a significant performance improvement was noted. The EHS cell voltages on Parsons' SRDFG using the G86-14-3 anode catalyst were 14 mV less on average, than those for the heat-treated Pt/Vulcan XC-72. In addition, the cell was able to operate with stable performance at up to 800 mA/cm² at 230°C and up to 1000 A/cm² at 250°C. At low current densities, 200 and 400 mA/cm², reasonable performance was achieved at 210°C. The data in Table 5 was gathered from Cell HS-114. This performance was repeated with an equivalent cell, HS-117, with comparable results. The product gas at the cathode contained 0.2% CO, 0.5% $CO_2$ and 6 ppm $H_2S$ at 600 mA/cm² and 250°C.

## TABLE 5

EHS cell voltages using the G86-14-3 anode electrocatalyst. Uncorrected cell voltage on pure $H_2$ and on Parsons' SRDFG at 70% $H_2$ utilization rate.

| Cell Temp., °C | Current Density mA/cm² | Cell Voltage, mV | | |
| --- | --- | --- | --- | --- |
| | | $H_2$ | SRDFG | $\triangle V$ |
| 210 | 200 | 41 | 59 | 18 |
| | 400 | 81 | 122* | 41 |
| | 600 | 118 | --- | -- |
| | 800 | 159 | --- | -- |
| | 1000 | 196 | --- | -- |
| Cell Resistance Correction Factor (ohm-cm²) | | 0.15 | 0.16 | |
| 230 | 200 | 43 | 57 | 14 |
| | 400 | 83 | 102 | 19 |
| | 600 | 125 | 152 | 27 |
| | 800 | 167 | 199 | 32 |
| | 1000 | 208 | --- | -- |
| Cell Resistance Correction Factor (ohm-cm²) | | 0.16 | 0.16 | |
| 250 | 200 | 47 | 66 | 19 |
| | 400 | 93 | 113 | 20 |
| | 600 | 135 | 164 | 29 |
| | 800 | 184 | 218 | 34 |
| | 1000 | 223 | 260 | 37 |
| Cell Resistance Correction Factor (ohm-cm²) | | 0.17 | 0.16 | |

*Voltage unstable and rising

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This catalyst powder was heat-treated at 900°C in $N_2$. The average particle size, 45 Å, was determined by transmission electron microscopy of the as-prepared powder. The pre-test surface area, as determined by hydrogen adsorption was 71 $m^2/g$, slightly higher than the 900°C heat-treated 10% Pt/Vulcan catalyst. The post-test surface area of Cell HS-114 was 74 $m^2/g$, slightly larger than the pre-test value. This increase is within experimental error and, therefore, is not significant. The post-test cathode showed only a small decrease in surface area, from 116 $m^2/g$ to 92 $m^2/g$. Catalyst sintering may have been limited by reduced time of operation at high temperatures for this cell. Only nine polarization scans were run on this cell at 230°C or above. Using an equivalent cell, HS-117, a decrease in surface area from 80 $m^2/g$, pre-test to 49 $m^2/g$, post-test was measured after fifteen polarization scans at 230°C or above.

The pre-test and post-test anodes from Cell HS-117 were examined by transmission election microscopy. The supported catalyst particles scraped from these electrodes are shown in Figures 3 and 4. The catalyst particle size had increased from 45 Å to approximately 100 Å. Agglomeration of several smaller particles into a larger particle is also evident.

Testing for the shift reaction or direct CO oxidation was performed by running Cell HS-117 on a $CO/H_2O/N_2$ feed at 230°C and 250°C. The $CO_2$ concentration in the anode exit stream increased from about 3% at 80 $mA/cm^2$ to about 12% at 200 $mA/cm^2$ while the CO content decreased from 8% to 1%. However, the cell voltage increased from 550-600 mV at 80 $mA/cm^2$ to over 700 mV at 200 $mA/cm^2$.

### 3.4 Catalyst G86-14-4

The results of testing this anode catalyst are shown in Table 6. Cell polarizations were higher than those seen with the G86-14-3 catalyst. Cell voltage on pure $H_2$, after iR-correction, was only slightly higher than that measured for the previous cell, but the iR-corrected cell voltage on Parsons' SRDFG was 1.5 to 2

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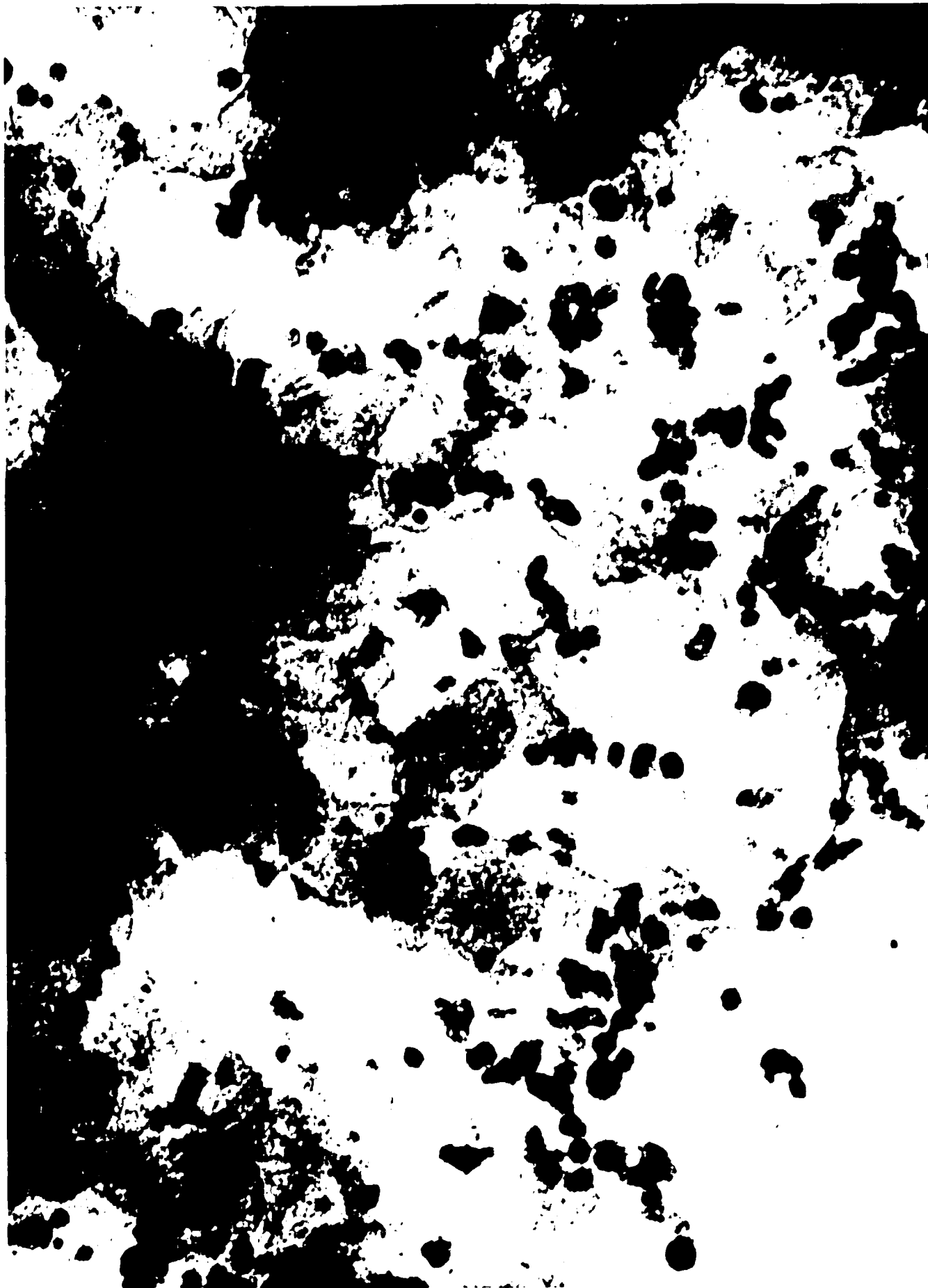
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Figure 5.  Catalyst Expelled from Pre-Test Anode.

Figure 4. Catalyst 386-14-3 From Post-Test Anode. 460,000X

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times larger with this anode catalyst. The cathode exit gas contained 0.9% CO, 1.5% $CO_2$ and 20 ppm $H_2S$ at 200 mA/cm² and 250°C. These contaminant levels are slightly higher than those measured at 600 mA/cm², 250°C in the cells discussed previously. Although higher contaminant concentrations might be expected at lower current densities, where less $H_2$ is produced at the cathode, no systematic relationship between contaminant concentrations and operating conditions could be discerned in any of the cell configurations.

TABLE 6

EHS cell voltages using the G86-14-4 formulation. Uncorrected cell voltage on pure $H_2$ and on Parsons' SRDFG at 70% $H_2$ utilization rate.

| Cell Temp., °C | Current Density mA/cm² | Cell Voltage, mV | | |
|---|---|---|---|---|
| | | $H_2$ | SRDFG | $\Delta V$ |
| 230 | 200 | 70 | 102 | 32 |
| | 400 | 140 | 194 | 54 |
| | 600 | 208 | 300 | 92 |
| | 800 | 273 | ---* | -- |
| | 1000 | 340 | --- | -- |
| Cell Resistance Correction Factor (ohm-cm²) | | 0.27 | 0.27 | |
| 250 | 200 | 78 | 107 | 29 |
| | 400 | 156 | 191 | 35 |
| | 600 | 226 | 277 | 51 |
| | 800 | 300 | 380 | 80 |
| | 1000 | 377 | 462 | 85 |
| Cell Resistance Correction Factor (ohm-cm²) | | 0.28 | 0.27 | |

*Voltage unstable and rising

The pre-test anode surface area, 68 m²/g, had decreased to 16 m²/g after EHS testing. The cathode surface area, however, only decreased from 116 m²/g to 73 m²/g. The large decrease in anode surface area may have been due to loss of catalyst. This is consistent with the extremely high voltages, high cell resistance and low $H_2$ output measured at the end of testing of this cell. Analysis of the pre-test and post-test anode electrodes by transmission electron microscopy also suggests that loss of catalyst contributed to the failure of this cell. The post-test

anode showed a very light loading of catalyst particles on the carbon support material, much lighter than seen on the pre-test anode. The pre-test catalyst particle size was the same as the G86-14-3 catalyst and the heat-treated Pt/Vulcan XC-72; approximately 40-60 Å.

The shift reaction or direct CO oxidation was noted in this cell at voltage levels similar to those seen in previous cells. At 230°C, the $CO_2$ content of the anode exit gas increased from 5% at 80 $mA/cm^2$ to 10% at 200 $mA/cm^2$ while the CO content decreased from 9% to 2%. Cell voltage was approximately 570 mV at 80 $mA/cm^2$ and rose to 770 mV at 200 $mA/cm^2$.

## 3.5   Catalyst G86-14-5

The EHS cell voltages using the G86-14-5 anode catalyst are shown in Table 7. Comparison of the cell voltages on pure $H_2$ of this catalyst and the other catalysts (Tables 3-6) shows that this catalyst performed better than the non-heat-treated Pt/Vulcan and the G86-14-4 catalyst, but slightly worse than the G86-14-3 catalyst. The cell voltages on pure $H_2$ of the G86-14-5 were very similar to those of the cell with a 900°C heat-treated Pt/Vulcan anode catalyst. However, operation on the Parsons' SRDFG resulted in extremely high cell voltages at current densities of 400 $mA/cm^2$ or higher. Although this catalyst was expected to show tolerance to CO, the $H_2S$ content of the SRDFG may have been responsible for this severe poisoning effect.

The cathode exit gas contained 0.2% CO, 0.2% $CO_2$ and a trace quantity of $H_2S$ at 200 $mA/cm^2$, 250°C. The pre-test anode surface area, 74 $m^2/g$, decreased to 54 $m^2/g$ after testing. The cathode surface area decreased from 121 $m^2/g$ to 36 $m^2/g$. TEM examination of scraping from pre-test and post-test anodes indicated that the loss of anode surface area was due, in part, to an increase in particle size. The pre-test catalyst particle size was approximately 35 Å. The post-test particle size was approximately 58 Å. This increase in particle size was not as large as that seen on the G86-14-3 post-test anode. However, the density of particles or catalyst

loading appeared lighter on the G86-14-5 post-test anode than on the G86-14-3 post-test anode. Some of the decrease in surface area of the G86-14-5 may have been due to loss of catalyst as well as catalyst agglomeration and sintering.

## TABLE 7

EHS cell voltages using the G86-14-5 anode catalyst. Uncorrected cell voltages on pure $H_2$ and on Parsons' SRDFG at 70% $H_2$ utilization rate.

| Cell Temp., °C | Current Density mA/cm$^2$ | Cell Voltage, mV | | |
| --- | --- | --- | --- | --- |
| | | $H_2$ | SRDFG | $\Delta V$ |
| 230 | 200 | 52 | 74 | 22 |
| | 400 | 104 | 230 | 126 |
| | 600 | 151 | 590 | 439 |
| | 800 | 202 | 890* | 688 |
| | 1000 | 250 | --- | --- |
| Cell Resistance Correction Factor (ohm-cm$^2$) | | 0.18 | 0.18 | |
| 250 | 200 | 56 | 73 | 17 |
| | 400 | 112 | 152 | 40 |
| | 600 | 161 | 375 | 214 |
| | 800 | 214 | 735 | 521 |
| | 1000 | 266 | --- | --- |
| Cell Resistance Correction Factor (ohm-cm$^2$) | | 0.18 | 0.18 | |

*Voltage unstable and rising

Testing for the shift reaction or direct CO oxidation revealed an increase in $CO_2$ content from 4% at 80 mA/cm$^2$ to 7.5% at 160 mA/cm$^2$. The CO content decreased from 9% to 1.5%. The voltage required rose from 498 mV at 80 mA/cm$^2$ to 740 mV at 160 mA/cm$^2$. Performance was unstable above 160 mA/cm$^2$. These results are poorer than seen with the G86-14-3 anode catalyst both in terms of voltage and amount of CO converted to $CO_2$.

## 3.6 Catalyst G86-14-6

This catalyst did not show good performance in the EHS cell. Cell voltages on pure $H_2$ and on Parsons' SRDFG are shown in Table 8. Cell voltages were generally higher in this cell than in

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the cell with the G86-14-5 anode catalyst. Performance on Parsons'
SRDFG was possible only at 250°C, with extremely high cell
polarization.

## TABLE 8

EHS cell voltages using the G86-14-6 anode catalyst.
Uncorrected cell voltage on pure $H_2$ and on Parsons' SRDFG at
70% $H_2$ utilization rate.

| Cell Temp., °C | Current Density mA/cm$^2$ | Cell Voltage, mV $H_2$ | SRDFG | $\Delta V$ |
|---|---|---|---|---|
| 230 | 200 | 61 | | |
| | 400 | 122 | | |
| | 600 | 178 | | |
| | 800 | 240 | | |
| | 1000 | 295 | | |
| Cell Resistance Correction Factor (ohm-cm$^2$) | | 0.19 | | |
| 250 | 200 | 74 | 98 | 24 |
| | 400 | 145 | 240 | 95 |
| | 600 | 225 | 435 | 210 |
| | 800 | 320 | 620* | 300 |
| | 1000 | 440 | ---* | --- |
| Cell Resistance Correction Factor (ohm-cm$^2$) | | 0.23 | 0.23 | |

*Voltage unstable and rising

Measurement of product gas composition and testing for the
shift reaction were not performed on this cell, as performance on
SRDFG was extremely poor. Pre and post-test anode surface areas
were 51 and 48 m$^2$/g, respectively. All of the other heat-treated
catalysts had pre-test surface areas of 60 m$^2$/g or more. While
this result indicates that poisoning of the anode surface or loss
of catalyst during testing are not responsible for the poor
performance, low surface area may have been a cause. The cathode
surface area dropped from 121 m$^2$/g to 30 m$^2$/g. The reason for this
large decrease in cathode surface area is unclear.

TEM analysis of the G86-14-6 catalyst, as prepared showed a
particle size of 58 Å for the precious metal alloy component. As
the catalyst particle size was significantly larger than the

particle size of the G86-14-3 or G86-14-5 catalysts (35-36 Å), a second preparation of the catalyst was made. The particle size, as measured by TEM, of the second preparation was 42 Å. The surface area of a pre-test anode manufactured from this second preparation as measured by hydrogen adsorption was 86 $m^2/g$. These values are similar to the results achieved with the other heat-treated catalysts.

However, when the second preparation of G86-14-6 was formed into an electrode, extremely high voltages at low current densities were measured on two subsequent cells. Further analysis of the reasons for this poor performance was not pursued due to time limitations.

## 4.0  CONCLUSIONS AND RECOMMENDATIONS

### 4.1  Conclusions

An electrochemical hydrogen separator has been developed which can produce essentially pure hydrogen from heavily contaminated simulated reformed diesel fuel gas. The EHS device operates by oxidizing the hydrogen content of the fuel gas to hydrogen ions at a gas-diffusion anode. The hydrogen ions are electrochemically transported across an electrolyte matrix to a gas diffusion cathode where they are recombined into hydrogen gas. The product gas contains only small quantities of CO, $CO_2$ and $H_2S$ which diffuse through the electrolyte matrix to the cathode. No systematic relationship between contaminant concentrations in the product gas and operating conditions or anode catalyst could be discerned.

An essential component of this device is an anode catalyst which can efficiently oxidize hydrogen in the presence of high levels of CO and $H_2S$. Efficient operation was highlighted by low operating voltages on $H_2$ and on SRDFG. Low polarization, the voltage difference between operation on pure $H_2$ and on SRDFG, typified an anode catalyst which resisted poisoning by CO and $H_2S$ and was tolerant to dilution of the hydrogen by $N_2$ and $CO_2$.

Six anode catalysts supported on Vulcan XC-72 were tested: 10% Pt, 10% Pt heat-treated at 900°C in $H_2$, and four proprietary formulations. The best performance was achieved with a proprietary formulation, G86-14-3. An operating voltage of 57 mV (25 mV after iR-correction) was measured at 230°C, 70% utilization, 200 mA/cm² during operation on SRDFG based on the output of the Ralph M. Parsons Co. reformer. This was only 14 mV above the operating voltage on pure $H_2$. Good performance was also achieved at current densities up to 800 mA/cm² at 230°C, up to 1000 mA/cm² at 250°C and up to 400 mA/cm² at 210°C. The product gas contained only 0.2% CO, 0.5% $CO_2$ and 6 ppm $H_2S$ at a current density of 600 mA/cm² and a temperature of 250°C.

The cell polarization generally increased with increasing current density and with increasing $H_2$ gas utilization in all cells. The effect of cell temperature was less clear. During operation on pure $H_2$ and operation on Parsons' SRDFG, the cell voltage generally increased with increasing temperature. During operation on Parsons' SRDFG, the cell voltages were generally lowest at 230°C. Slight increases in voltage were seen at 250°C and 210°C (stable operation at 210°C was possible only with the G86-14-3 catalyst).

Testing for the shift reaction or direct CO oxidation was also performed in order to determine if the high CO content of the SRDFG could be used to generate additional hydrogen through oxidation of CO to $CO_2$ with $H_2O$. The EHS cell was operated on a $CO/N_2/H_2O$ feed at 230°C and 250°C. $CO_2$ and $H_2$ were produced in increasing amounts at current densities from 80 mA/cm² to 160-200 mA/cm². However, the cell voltage required also increased with current density from 500-600 mV to 650-770 mV; the exact values varied depending on the anode catalyst.

TEM analysis of as-prepared catalyst powders and pre-test and post-test electrodes revealed the effect of cell operation on catalyst particle size and loading. Significant increases in particle size were measured, likely due to agglomeration and sintering. In the case of the G86-14-3 catalyst, the average

particle size more than doubled, from 45 Å to 100 Å. However, this did not seem to seriously degrade the cell performance. Poor performance may be related to a loss of catalyst particles during testing, as was seen in post-test analysis of the G86-14-4 and G86-14-5 electrodes.

Pre-test and post-test catalyst surface area was determined by hydrogen adsorption via cyclic voltammetry. In general, the post-test surface areas decreased, relative to the pre-test surface areas. However, a large decrease in surface area could not be directly related to poor performance. The amount of testing done at 230°C and above did appear to relate to the decrease in surface area. Cells which were not tested extensively at high temperatures, e.g., nine or fewer polarization scans at 230°C and above, showed little decrease in anode surface area. The majority of the anodes showed a post-test surface area of approximately 50 m$^2$/g after extensive testing.

In conclusion, the G83-14-3 material has been identified as the best anode catalyst for the EHS device. This catalyst shows excellent tolerance to high levels of CO and $H_2S$ present in Parsons' SRDFG. The EHS cell was capable of producing high quality, pure $H_2$ with minimal voltage requirement. Operation at 210, 230 and 250°C was achieved at maximum current densities of 400, 800, and 1000 mA/cm$^2$, respectively, with 230°C being identified as the optimum operating temperature. Use of the EHS device with this anode catalyst would allow pure hydrogen gas for use in the PAFC to be generated from fuel gases with increased contaminant concentrations. This would allow reduction in size and weight of the shift convertor component of the fuel processing train. The shift convertor represents approximately 7% of the total system weight of a 150 kW PAFC power plant. Complete elimination of the shift convertor is probably not feasible since a large portion of the energy value of the SRDFG is in the CO content of the gas. An increase in post-shift convertor CO content from 2% to 5%, however, would allow a reduction in the size and weight of the shift convertor by approximately 10 to 20%. Tolerance to CO levels well above 5% has been demonstrated in the EHS using the advanced anode

catalyst developed in this program. The tolerance to high levels of $H_2S$ in the EHS cell would also reduce the gas cleanup requirements in other components of the fuel processing train. For instance, the requirements for the zinc oxide guard bed would be greatly reduced or eliminated, resulting in additional reduction of the fuel processor size and weight. These weight and volume savings would be realized at minimal loss in overall voltage output of the PAFC power generation system due to the efficiency of the EHS device.

## 4.2  Recommendations

Several directions for future work are suggested by the results of this investigation. The catalyst system G86-14-3 identified in this program merits further study. Modifications of this formulation may provide comparable performance at a wider range of temperatures, i.e., above 250°C and/or below 210°C. Additions to the catalyst system may be developed which would encourage the shift reaction or CO oxidation to take place at the EHS anode, thus utilizing the available energy from the CO. This would further reduce the need for the shift convertor components of the fuel processing train. This investigation has focused on the use of reformed diesel fuel gas as a hydrogen source for the phosphoric acid fuel cell. The EHS device has shown promise for making this feasible. This approach could easily be transferred to allow the use of other hydrogen-containing fuel gases in the PAFC, such as coal gas. Coal gas can have a wide variety of composi-tions, but CO and $H_2S$, two of the major contaminants, are present in amounts similar to that seen in the SRDFG used here. Lastly, this technology may be transferred to use in the PAFC anode so that the PAFC can operate directly on contaminated gases. This would likely require extensive testing and development to determine the low temperature tolerance, long-term stability and performance under pressurized operation. The motivation for this work is provided by expected reductions of size and cost in the fuel processing components of a PAFC power generation system.

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